Abstract of B1

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(54) METHOD FOR ADJUSTING EVAPORATION RATE OF AROMATIC COMPOUND BY UTILIZATION OF CRYSTALLIZATION CHARACTERISTIC WITH SURFACTANT

(57)Abstract:
PROBLEM TO BE SOLVED: To provide a method for simply controlling the evaporation of a fragrant, a medicine or the like at low cost to impart a controlled release property.
SOLUTION: This method for adjusting the evaporation rate of the aromatic compound, comprising the formation of the crystals of the molecular complex of the aromatic compound with a surfactant.

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* NOTICES *

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CLAIMS

[Claim(s)]
[Claim 1] How to control the evaporation rate of an aromatic compound by forming the crystal of the molecule complex of a surfactant and an aromatic compound.
[Claim 2] a with a carbon numbers [of at least 1 which said surfactant combined with the radical and this radical of ionicity] of eight or more straight chain alkyl group -- since -- the approach according to claim 1 characterized by being the becoming ionic surfactant.
[Claim 3] The approach according to claim 2 characterized by adjusting an evaporation rate with the die length of the chain of the alkyl group of said surfactant.
[Claim 4] The approach according to claim 1 to 3 characterized by being chosen from the group which said ionic surfactant becomes from the sulfate of quarternary ammonium salt, alkylamine salt, alkyl pyridinium salt, an alkyl quinolinium salt, an alkyl iso quinolinium salt, a fatty-acid salt, fatty alcohol sulfate, a liquid fatty-oil sulfate salt, fatty amine, and an aliphatic series amide, fatty alcohol phosphate, the sulfonate of dibasicity fatty acid ester, a fatty-acid amidosulfonic acid salt, and alkylaryl sulfonates.
[Claim 5] The approach according to claim 1 to 4 said aromatic compound is perfume.
[Claim 6] The approach according to claim 1 to 4 said aromatic compound is a biocide.
[Claim 7] The approach according to claim 1 to 4 said aromatic compound is physic.
[Claim 8] the ionic surfactant which consists of a radical of the ionicity combined with the with a carbon numbers of eight or more alkyl group and this alkyl group of at least 1, and an volatile aromatic compound -- since -- the becoming molecule complex crystal.

[Translation done.]

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DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[Field of the Invention] This invention relates to the approach of adjusting the evaporation rate of an aromatic compound, by forming the molecule complex crystal of a surfactant and an aromatic compound, and this crystal. By using this crystal and approach, the evaporation rate of an volatile aromatic compound can be adjusted cheaply and easily, and chemicals or perfume, such as physic, agricultural chemicals, an insecticide, a parasiticide, a germicide, a disinfectant, and cosmetics, etc. can be saved at stability, or it can give them sustained-release.
[0002]
[Description of the Prior Art] In order that a non-portable aromatic and a non-portable insecticide may generally control the evaporation rate, What dissolved perfume, or sterilization and an insecticidal component in the liquid etc. is put into the container of an open sand mold. Or long-term its durability and sustained-release one are maintained by putting in in the container which made opening the configuration which can suppress evaporation of an active ingredient, installing indoors etc., these active ingredients' evaporating gradually, and the gas of an active ingredient being made to be emitted into atmospheric air.
[0003] Moreover, as an option which controls sustained-release [of an aromatic or an insecticide / evaporation or sustained-release], infiltrate the liquid which dissolved the active ingredient in other solid matter, the active ingredient itself is made to react with other compounds, or there is the approach of forming a clathrate compound. On the other hand also in the field of physic or cosmetics, the drugs which demonstrate the gradual release operation which can maintain that effectiveness by one spreading for a long time are needed in external preparations or cosmetics applied to the skin, for example, and the porous film and the hydrogel are used for this purpose. Furthermore, also, for example in inhalations, the drugs which have a gradual release operation for the purpose of simple and prolonged repetitive administration etc. are demanded.
[0004] However, even if it is which [above-mentioned] case, in order to control evaporation or to give a gradual release operation, a compound, equipment, etc. of the specification as which the gradual release operation concerned is required peculiar to an activity compound were required, manufacture took time and effort, and there was a fault that cost also became high. Moreover, there was a fault that it was difficult to adjust extent of the evaporation / gradual release operation.
[0005]
[Problem(s) to be Solved by the Invention] Therefore, the purpose of this invention is developing the approach evaporation of compounds, such as perfume, and an insecticide, physic, being controlled simple and cheaply, or the evaporation / gradual release operation being adjusted easily [can give sustained-release and].
[0006]
[Means for Solving the Problem] That such a problem should be solved, as a result of repeating research wholeheartedly, this invention person found out that the molecule complex crystal formed between the surfactant and the aromatic compound gave prevention or sustained-

release for evaporation of said aromatic compound, and could adjust the evaporation rate easily, and completed this invention based on this.

[0007] this invention persons showed clearly that a surfactant can form a crystal as a molecule complex with various aromatic compounds, and have succeeded in the spacial configuration analysis of this molecule complex crystal by X-ray crystallographic analysis before (Bull.Chem.Soc.Jap., 71, and 2109-2118 (1998)). The invention in this application is based on the knowledge that this formed molecule complex crystal can control the volatility of an aromatic compound.

[0008] Therefore, this invention relates to the approach of adjusting the evaporation rate of an aromatic compound, by forming a molecule complex crystal in the molecule complex crystal and list which were formed between the surfactant and said aromatic compound between a surfactant and said aromatic compound. In addition, a "crystal" means what is solid matter which had periodic atomic arrangement spatially, and takes space lattice structure, and, unlike a micell, differs from suspension or an emulsion as generally recognized. Moreover, the crystal in which a "molecule complex crystal" says the crystal of a molecule complex to, and is formed with the surfactant and aromatic compound of a fixed presentation ratio is said.

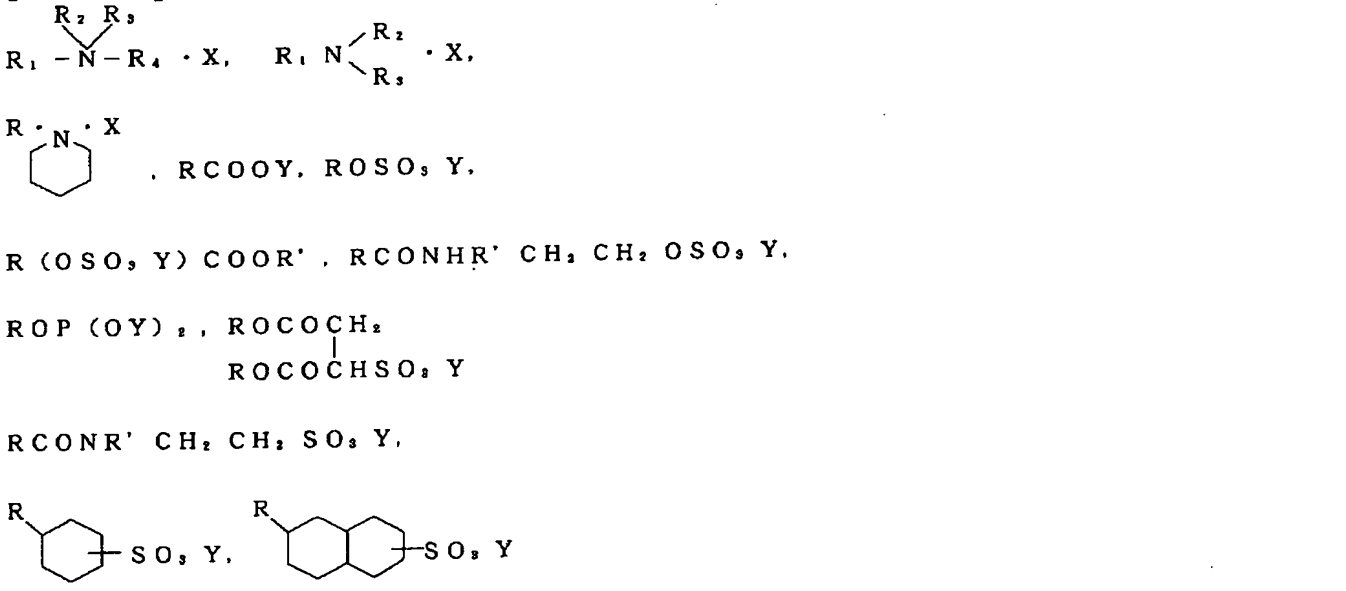
[0009]

[Embodiment of the Invention] A surfactant and an aromatic compound are used for the molecule complex crystal and approach by this invention. Although what will not be limited especially if an aromatic compound and a molecule complex crystal can be formed according to the purpose of this invention, but is generally marketed can be used, a surfactant needs to be the surfactant permitted physiologically, when using as physic, corresponding to the purpose. Moreover, as for a surfactant, it is desirable that it is the ionic surfactant which has the radical of ionicity from the result of structural analysis of a publication below. An ionic surfactant may be dipolar ion nature also in anionic or cationicity.

[0010] As an example of a surface active agent, quarternary ammonium salt, alkylamine salt and alkyl pyridinium salt, an alkyl quinolinium salt, an alkyl iso quinolinium salt, etc. have the sulfate of a fatty-acid salt, fatty alcohol sulfate, a liquid fatty-oil sulfate salt, fatty amine, and an aliphatic series amide, fatty alcohol phosphate, the sulfonate of dibasicity fatty acid ester, a fatty-acid amidosulfonic acid salt, alkylaryl sulfonates, etc. as an anionic surface active agent as a cationic surface active agent.

[0011] Specifically, it is following surfactant: [0012].

[Formula 1]



(R1, R2, R3, and R4 are R, R', and the alkyl group permuted by hydrogen or arbitration respectively independently among a formula, X is a halogen, for example, chlorine, or a bromine, and Y is alkali metal, for example, sodium, or a potassium) is desirable.

[0013] Especially the classes of desirable surface active agent are an alkyl trimethylammonium salt, a dialkyl dimethylannmonium salt, an alkyl dimethylbenzyl ammonium salt, alkyl pyridinium salt, a fatty-acid monocarboxylic acid salt, alkylbenzene sulfonates, sulfo succinic-acid dialkyl ester, an alkyl sulfate salt, an alkyl sulfate polyoxyethylene salt, and a phosphoric-acid alkyl salt.
[0014] Moreover, as for viewpoints, such as packing of the result of structural analysis by experiment given in the following to a molecule complex crystal, formation of the space in a crystal, and an interaction with an aromatic compound, to a surfactant, it is desirable about an above-mentioned alkyl group to have what added the permutation by the hydrophobic radical to 1 of a with a carbon numbers [of at least 1] of eight or more straight chain, a branched chain alkyl group, or this alkyl group or two or more places. Preferably, a surfactant has a with a carbon numbers [of at least 1 combined with the radical of said ionicity] of eight or more straight chain alkyl group. Especially an alkyl group is an octyl radical, a nonyl radical, a decyl group, an undecyl radical, the dodecyl, a tridecyl radical, a tetradecyl radical, a pentadecyl group, a hexadecyl radical, a heptadecyl radical, an octadecyl radical, a nona decyl group, or an icosyl group. the carbon number of a desirable alkyl group -- 8-30 -- more -- desirable -- 8-20 -- it is 10-16 most preferably.
[0015] However, association becomes firm by that in a crystal, so that from the experiment of a publication and the carbon number of this alkyl chain becomes large. [below] Therefore, an aromatic compound becomes stability more and the evaporation rate decreases. Therefore, sustained-release [which is demanded about a predetermined aromatic compound / the evaporation or sustained-release] can determine the carbon number of this alkyl group freely. Moreover, thereby, it can acquire desired evaporation rate and sustained-release one.
[0016] Especially a desirable surface active agent is the halogenide of DESHIRU trimethylammonium, dodecyl trimethylammonium, tetradecyl trimethylammonium, or hexadecyl trimethylammonium. It may be perfume, or may be a biocide, or you may be physic, control of the evaporation rate, i.e., sustained-release, shelf life, etc. are required, and the aromatic compound which forms a surfactant and a crystal will not be especially limited, if it is the compound which can form a crystal as a molecule complex with a surfactant. Here, a biocide is used in order to eliminate or kill a harmful living thing especially including an insecticide, miticide, a germicide, a fungicide, a parasiticide, a disinfectant, etc.
[0017] However, the aromatic compound with which an aromatic compound has less than three rings or heterocycles in all in consideration of packing, an interaction with a surfactant, occupancy space, etc. of the result of structural analysis by experiment given in the following to a molecule complex crystal is desirable. Preferably, these rings are mostly located in a line on a straight line. desirable -- the molecular weight of an aromatic compound -- 78-300 -- more -- desirable -- 78-200 -- it is 78-150 most preferably. desirable -- the carbon number of an aromatic compound -- 6-30 -- more -- desirable -- 6-20 -- it is 6-10 most preferably.
[0018] When a concrete example is given, an aromatic compound o-iodine phenol, m-iodine phenol, p-iodine phenol, p-cresol, m-cyano phenol, o-hydroxybenzoic acid, m-hydroxybenzoic acid, para hydroxybenzoic acid, ortho toluylic acid, Meta toluylic acid, para toluylic acid, o-phthalic acid, meta phthalic acid, para phthalic acid, A hydroquinone, 1, 4-dimethoxybenzene, para benzoquinone, 1, 4-cyclohexane diol, Naphthalene, 1-naphthol, 2-naphthol, Indore, 7-hydroxycoumarin, A coumarin, 2-aminopyridine, 3-aminopyridine, 4-aminopyridine, 2-hydroxypyridine, a 3-hydroxy pyridine, a 4-hydroxy pyridine, 3-cyano torr pyridine, 4-SHIANOSHI pyridine, 4-dimethylaminopyridine, 2-phenyl pyridine, 3-phenyl pyridine, 4-phenyl pyridine, A - bipyridine, 2, 4-bipyridine, and 2 and 2 '4, 4'-bipyridine, An anthracene, an acridine, a phenanthrene, and benzo[h] Quinoline, Benzo[f] Quinoline, 1, and 1'-biphenyl-4-oar, dibenzyl, It is especially chosen from a biphenyl, a carbazole dibenzofuran and a diphenylamine, and flopropion and 4-chloro-m-cresol and the group that consists of guaiacol, 2-methylindole, and skatole as perfume as physic.
[0019] The molecule complex crystal by this invention can be manufactured by the method of crystallizing common use, for example, it can dissolve a surfactant and an aromatic compound in a suitable solvent by the suitable mole ratio, can be settled by leaving it at suitable temperature, and can be easily manufactured by isolating this. Moreover, the molecule complex crystal by this

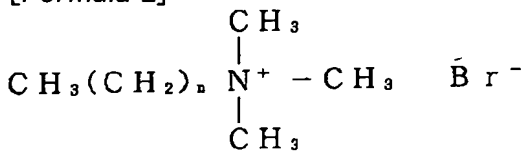
invention can be made only by mixing these solid-states in a mortar etc. depending on a surfactant and an aromatic compound. When this carries out X-ray crystallographic analysis of the product, it is checked that it is the crystal of a molecule complex.

[0020] The approach and crystal of this invention can control evaporation of an aromatic compound using a cheap and very common surfactant. Therefore, it can use in order to save the agent or drugs like physic, an insecticide, a parasiticide, a germicide, a disinfectant, or an aromatic or to give them a gradual release operation.

[0021]

[Example] It sets to the following experiments and is hexadecyl trimethylammonium bromide (CTAB; Wako), tetradecyl trimethylammonium bromide (MTAB; Wako), dodecyltrimethylammonium bromide (LTAB; Tokyo formation), and DESHIRU trimethylammonium bromide (DTAB; Tokyo formation): [0022] as a surface active agent.

[Formula 2]

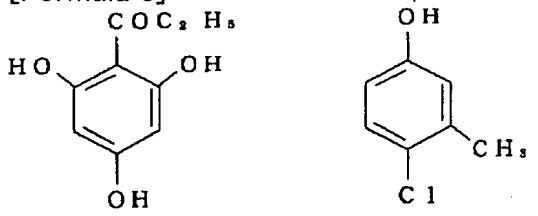


n＝15 ヘキサデシルトリメチルアンモニウムブロミド
n＝13 テトラデシルトリメチルアンモニウムブロミド
n＝11 ドデシルトリメチルアンモニウムブロミド
n＝9 デシルトリメチルアンモニウムブロミド

********.

As an aromatic compound used as crystal 1. ingredient physic of a <example 1> surfactant and a physic compound, it is flopropion (antispasmodic) (IGMA) and 4-chloro-m-cresol (germicide) (Tokyo formation): [0023].

[Formula 3]



フロプロピオン　　4－クロロ－m－クレゾール

********. These used for molecule complex crystallization that which recrystallized by the approach of common use. CTAB and MTAB were respectively crystallized from the methanol-acetone solution, then, it recrystallized from water, and this was used for the following molecule complex crystallization.

[0024] 2. After having added the 4-chloro-m-cresol or flopropion of an equimolecular amount to the methanol solution including the preparation CTAB and MTAB of a molecule complex crystal respectively, warming by the water bath and making it a homogeneity solution, the precipitate which left it in the cool place for about one week, and was formed in it was isolated, and two molecule complex crystal 1-I and 1-II(s) were obtained.

[0025] After solubilizing until it added the flopropion of an equimolecular amount in the water solution containing CTAB using the usual solubilizing method and obtained the homogeneity solution, the precipitate which left it in the cool place for about one week, and was formed in it was isolated, and one molecule complex crystal 1-III was obtained. After the obtained molecule complex crystal fully dried these, it dissolved in the methanol solution, measured absorption wavelength using spectrophotometer for ultraviolet and visible region (UV160A, Shimadzu), and checked formation of a molecule complex by comparing this value with the absorption wavelength of a simple substance.

[0026] Each of crystal 1-I, 1-II(s), and 1-III(s) was non-color plate-like crystals.
3. X-ray crystallographic analysis was performed about X-ray-structural-analysis crystal 1-I of a molecule complex crystal, 1-II, and 1-III. The crystal used the X-ray which MoKalpha or CuKalpha monochrome-ized using the CCD DI contact sense non-isolated (SMART-CCD;Simens) or the 4 shaft type automatic DI contact sense non-isolated (AFC-5 R;Rigaku) after cooling at -50 degrees C using the nitrogen spraying mold cooling system. SADABS or psi-scan is used for absorption amendment, and it is program SIR-92[111]. It uses, a phase is determined according to a direct method, and it is least square method program SHELXL-97 [109]. Elaboration was carried out. Each experiment conditions and crystallographic data are shown in the following table 1.
[0027]
[Table 1]

表1 結晶学的データと測定条件

		(1-I)	(1-II)	(1-III)
化学式		$C_{19}H_{42}NBr$ /0.5C_7H_7OCl	$2C_{17}H_{38}NBr$ /$C_9H_{10}O_4$	$2C_{19}H_{42}NBr$ /$2C_9H_{10}O_4$/$3H_2O$
分子量		435.73	854.96	1147.28
温度／K		223	223	223
波長／Å		0.71069	0.71069	1.54180
回折計		SMART-CCD	SMART-CCD	AFC-5R
放射線		MoKα	MoKα	CuKα
晶系		monoclinic	monoclinic	triclinic
空間群		P21	P21/m	P1
a／Å		5.5232(1)	11.6511(6)	9.747(2)
b／Å		7.3944(1)	7.3421(4)	40.114(16)
c／Å		33.3594(3)	28.8002(14)	9.214(2)
α／°		90	90	93.80(3)
β／°		95.058(1)	99.491(1)	117.869(17)
γ／°		90	90	90.41(3)
Z		2	2	2
体積／Å^3		1357.12(2)	2429.9(2)	3174.8(16)
D_{calc}／g cm^{-3}		1.066	1.168	1.200
結晶の寸法／mm^3		0.28x0.25x0.04	0.30x0.25x0.08	0.40x0.35x0.07
吸収補正		SADABS	SADABS	psi-scan (0.8374<T<1.000)
2θmax／°		55	55	135
μ／mm^{-1}		1.571	1.705	2.041
F (000)		470	920	1236
hの範囲		-7→7	-14→15	0→11
kの範囲		-9→9	-8→9	-48→48
lの範囲		-43→43	-37→37	-11→9
反射数	Total	18276	17606	11079
	Unique	6223	6006	10387
精密化パラメータの数		268	302	624
R (int)		0.056	0.042	0.030
R (I>2σ)		0.068	0.042	0.060
wR (F^2)		0.179	0.123	0.170
適合度 (F^2)		1.003	0.971	1.035
荷重パラメータ	a	0.1355	0.0783	0.1318
	b	0	0	0
δp／e Å^{-3}		+1.66, -0.84*	+0.40, -0.49	+0.80, -0.67

*これらのピークはBrから1.1 Å以内である

Structural drawing of these crystals is shown in drawing 1 -6. Here, as for drawing 1 and 2, drawing 3 and 4 are drawings in which drawing 5 and 6 show [I / crystal 1-] the crystal structure about crystal 1-III about crystal 1-II. CTAB of one molecule and the 4-chloro-m-cresol of 0.5 molecules existed in the unsymmetrical unit, in crystal 1-II, dyad MTAB and the flopropion of one molecule existed in the unsymmetrical unit, and, as for crystal 1-I, dyad CTAB and dyad flopropion existed [crystal 1-III] in the unsymmetrical unit. It is clear these [all] to form the molecule complex crystal.

[0028] 4. Solubility was measured in order to check formation of the measurement molecule complex of a dissolution rate. The mixture of a flopropion simple substance, and CTAB and flopropion powder and the molecule complex crystal of CTAB and flopropion were used as a sample. Each sample was sifted out by 40mesh(es) to 80mesh(es) and a degree, and it used for dissolution rate measurement. After having dissolved in distilled water of 500mL(s) which deaerated the sample under the condition of 25 degrees C and 50r.p.m. using dissolution rate measuring device U.S.P.(NF) NTR-5S3 (Toyama), extracting 10 mLs at a time for every fixed time amount and filtering using 0.45-micrometer membrane filter, the amount of dissolutions of flopropion was measured using spectrophotometer for ultraviolet and visible region (UV-160A, Shimadzu).

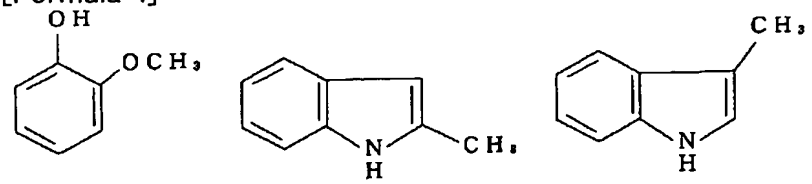
[0029] A result is shown in drawing 7 . From this drawing, the molecule complex crystal of CTAB and flopropion is understood that a dissolution rate is very quick compared with other two. This suggests existence of a molecule complex and suggests that contact in a flopropion molecule and the water which is a solvent is prevented. Moreover, it became clear that the further operation of improving the solubility of a water-insoluble nature aromatic compound also does so the molecule complex crystal by the invention in this application. This is advantageous in especially the application as physic.

[0030] 5. The gradual release operation over the 4-chloro-m-cresol of a molecule complex crystal was measured by measuring the decrement of the 4-chloro-m-cresol under molecule complex crystal accompanying a temperature rise in a 25-160-degree C temperature requirement by programming-rate 10 K/min under a nitrogen air current using the measurement RigakuTG8120 of a gradual release operation, and comparing this with the thing of a 4-chloro-m-cresol simple substance.

[0031] A result is shown in drawing 8 . This drawing shows that a molecule complex crystal controls evaporation of the drugs of a simple substance. Furthermore, it turns out that the evaporation rate is stopped more, so that the chain length of a surfactant is [the evaporation rate of the aromatic series molecule crystallized together] long depending on the die length of the chain of a surfactant. Furthermore, depending on the chain length of a surfactant, it turns out that the way when making it a surfactant and a molecule complex crystal is more more stable than the time of a drugs molecule simple substance.

As an aromatic compound used as a crystal 1. ingredient aromatic of a <example 2> surfactant and an aromatic compound, it is guaiacol (Tokyo formation), 2-methylindole (Aldrich), and skatole (Aldrich): [0032].

[Formula 4]



グアヤコール　　2-メチルインドール　　スカトール

********. Guaiacol used for molecule complex crystallization that which used the commercial item for molecule complex crystallization as it was since it was a liquid in ordinary temperature, was alike other than this, therefore recrystallized by the approach of common use. CTAB, MTAB, LTAB, and DTAB were respectively crystallized from the methanol-acetone mixed solution, then,

it recrystallized from water, and this was used for the following molecule complex crystallization.
[0033] In addition, when the mole ratio of these complexes was calculated using UV spectrum, it was set to mole-ratio =1:2 of a surfactant:aromatic compound also about which complex. 2. Preparation CTAB / guaiacol molecule complex crystal of a molecule complex crystal solubilized by having added the guaiacol of an equimolecular amount to CTAB water-solution 0.5x10-2mol/L, left the solution made into the homogeneity for one week in the 2-3-degree C cool place, isolated precipitate, and obtained molecule complex crystal 2-VII.
[0034] The molecule complex of the molecule complex of CTAB, MTAB or LTAB, and 2-methylindole and CTAB, MTAB, LTAB or DTAB, and skatole The perfume of it and an equimolecular amount was dissolved in the methanol, these surfactants were prepared to 0.5x10-2 mol/L, this was left for about one week at about 10 degrees C, precipitate was isolated, and molecule complex crystal 2-I, 2-II, 2-III, 2-IV, 2-V, and 2-VI were obtained.
[0035] After the obtained molecule complex crystal fully dried these, it dissolved in the methanol solution, measured absorption wavelength using spectrophotometer for ultraviolet and visible region (UV160A, Shimadzu), and checked formation of a molecule complex by comparing this value with the absorption wavelength of a simple substance.
3. X-ray crystallographic analysis was performed about X-ray-structural-analysis crystal 2-I-VII of a molecule complex crystal. The crystal used the X-ray which MoKalpha monochrome-ized using the CCD DI contact sense non-isolated (SMART-CCD;Simens) after cooling at -50 degrees C using the nitrogen spraying mold cooling system. Program SIR-92[111] It uses, a phase is determined according to a direct method, and it is least square method program SHELXL-97[109]. Elaboration was carried out. Each experiment conditions and crystallographic data are shown in the following tables 2-4.
[0036]
[Table 2]

表2　結晶学的データと測定条件

	(2－I)	(2－II)	(2－III)
化学式	$C_{19}H_{42}NBr$ /0.5C_8H_8N	$C_{17}H_{38}NBr$ /0.5C_8H_8N	$C_{15}H_{34}NBr$ /0.5C_8H_8N
分子量	430.03	401.98	373.93
温度／K	223	223	223
波長／Å	0.710969	0.71069	0.71069
回折計	SMART-CCD	SMART-CCD	SMART-CCD
放射線	MoKα	MoKα	MoKα
晶系	monoclinic	monoclinic	monoclinic
空間群	$P2_1$	$P2_1$	$P2_1$
a／Å	5.5160(16)	5.5240(1)	5.5400(1)
b／Å	7.4290(14)	7.4240(1)	7.4173(2)
c／Å	31.680(6)	29.5280(2)	27.3813(3)
α／°	90	90	90
β／°	90.449(11)	92.879(1)	95.786(1)
γ／°	90	90	90
Z	2	2	2
体積／Å^3	1298.2(5)	1209.42(3)	1119.42(4)
D_{calc}／g cm^{-3}	1.100	1.104	1.109
結晶の寸法／mm^3	0.25x0.25x0.10	0.30x0.20x0.10	0.30x0.20x0.08
吸収補正	SADABS	SADABS	SADABS
2θmax／°	55	55	55
μ／mm^{-1}	1.591	1.704	1.836
F (000)	466	434	402
hの範囲	-6→6	-7→6	-6→7
kの範囲	-9→9	-9→9	-9→9
lの範囲	-42→38	-38→38	-35→32
反射数 Total	9860	12612	7947
反射数 Unique	6026	5207	4699
精密化パラメータの数	271	205	228
R (int)	0.017	0.023	0.018
R (I＞2σ)	0.048	0.046	0.042
wR (F^2)	0.125	0.135	0.123
適合度 (F^2)	1.129	1.088	1.057
荷重パラメータ a	0.0676	0.0864	0.0835
荷重パラメータ b	0.86	0.88	0.48
δ p／e Å^{-3}	+0.93,-0.52	+1.19,-0.46	+1.07,-0.46

[0037]
[Table 3]

		(2－IV)	(2－V)
化学式		$C_{19}H_{42}NBr$ /0.5C_9H_9N	$C_{17}H_{38}NBr$ /0.5C_9H_9N
分子量		430.03	401.98
温度／K		223	223
波長／Å		0.71069	0.710969
回折計		SMART-CCD	SMART-CCD
放射線		MoKα	MoKα
晶系		monoclinic	monoclinic
空間群		$P2_1$	$P2_1$
a／Å		5.4943(3)	5.5092(12)
b／Å		7.4105(4)	7.395(2)
c／Å		32.3954(19)	30.204(6)
α／°		90	90
β／°		92.313(1)	94.325(18)
γ／°		90	90
Z		2	2
体積／Å^3		1317.92(13)	1227.0(5)
D_{calc}／g cm^{-3}		1.084	1.088
結晶の寸法／mm^3		0.25x0.25x0.20	0.22x0.20x0.08
吸収補正		SADABS	SADABS
2θmax／°		55	55
μ／mm^{-1}		1.567	1.679
F (000)		466	434
hの範囲		-6→7	-7→7
kの範囲		-9→9	-9→8
lの範囲		-34→42	-34→39
反射数	Total	7507	8687
	Unique	4601	4633
精密化パラメータの数		255	270
R (int)		0.017	0.022
R (I＞2σ)		0.060	0.060
wR (F^2)		0.166	0.152
適合度 (F^2)		1.070	1.069
荷重パラメータ	a	0.1003	0.0685
	b	1.68	2.74
δ p／e Å^{-3}		+1.92,-1.57	+1.78,-1.82

[0038]
[Table 4]

		(2-VI)	(2-VII)
化学式		$C_{15}H_{34}NBr$ /0.5C_9H_9N	$C_{19}H_{42}NBr$ /0.5$C_7H_8O_2$
分子量		373.93	426.51
温度／K		223	223
波長／Å		0.71069	0.71069
回折計		SMART-CCD	SMART-CCD
放射線		MoKα	MoKα
晶系		monoclinic	monoclinic
空間群		$P2_1$	$P2_1$
a／Å		5.5072(2)	5.5123(2)
b／Å		7.3926(2)	7.4272(3)
c／Å		27.9224(7)	32.4438(14)
α／°		90	90
β／°		94.068(1)	92.247(1)
γ／°		90	90
Z		2	2
体積／Å^3		1133.93(6)	1327.26(9)
D_{calc}／g cm^{-3}		1.095	1.067
結晶の寸法／mm^3		0.20x0.20x0.10	0.30x0.30x0.05
吸収補正		SADABS	SADABS
2θmax／°		55	55
μ／mm^{-1}		1.813	1.558
F (000)		402	462
hの範囲		-7→7	-6→5
kの範囲		-9→9	-8→7
lの範囲		-28→36	-35→32
反射数	Total	8047	6987
	Unique	4882	3585
精密化パラメータの数		237	262
R (int)		0.015	0.032
R (I>2σ)		0.059	0.048
wR (F^2)		0.152	0.154
適合度 (F^2)		1.106	1.086
荷重パラメータ	a	0.0617	0.1221
	b	2.63	0
δp／e Å^{-3}		+2.31, -1.52	+0.98, -0.59

Structural drawing of these crystals is shown in drawing 9 -15. Here, it reaches drawing 9 R>9, and as for 10, crystal 2-II, it reaches and crystal 2-I, drawing 11 , and 12 are drawings in which, as for crystal 2-VI, V and VI, and drawing 15 , III, drawing 13 , and 14 show the crystal structure about crystal 2-VII. It is clear these [all] to form the molecule complex crystal.

[0039] 4. The decrement of the aromatic compound under molecule complex crystal accompanying a temperature rise was measured in the 25-160-degree C temperature requirement by programming-rate 10 K/min under the nitrogen air current using the measurement RigakuTG8120 of a gradual release operation, and this was compared with the thing of an aromatic simple substance. The result of the experiment conducted on three sorts of molecule complex crystals, CTAB / 2-methylindole, MTAB / 2-methylindole, and LTAB/methylindole, and a list with 2-methylindole simple substance as contrast is shown in drawing 16 .

[0040] Furthermore, the result of the experiment conducted on four sorts of molecule complex crystals, CTAB/skatole, MTAB/skatole, LTAB/skatole, and DTAB/skatole, and a list with the

skatole simple substance as contrast is shown in drawing 17 . These drawings show that a molecule complex crystal controls evaporation of an aromatic depending on the die length of the chain of a surfactant like an example 1. Moreover, it is [at the time of forming a surfactant and a molecule complex] more more extremely stable than a simple substance, and it also turns out that it is dependent on the chain length. Namely, the molecule complex crystal by the invention in this application controls the evaporation rate of an aromatic compound, and, thereby, the application to physic, an aromatic, an insecticide, etc. is possible for it.

[0041] Furthermore, in order to prove the stability of these molecule complex crystal thermodynamically, Lennard-Jones potential is used using the data of a crystal structure, and it is van about each molecule complex. der waals energy was calculated. Count was performed only by the C-C interaction located in less than 5A between the alkyl chains of an adjacent surfactant. Here, since the location of a carbon atom asks for the hydrogen atom of the alkyl chain of a surfactant geometrically, it can be enough guessed only by count of a C-C interaction. Moreover, electrostatic energy was disregarded from the bromide anion and ammonium cation of a surface active agent occupying the almost same location in each crystal.

[0042] van of the molecule complex crystal formed in the following table 5 between CTAB, MTAB, LTAB or DTAB, 2-methylindole, or skatole der waals energy is shown.

[0043]

[Table 5]

表5 Lennard-Jones potentialを用いた分子錯体のvan der waalsエネルギーの算出

分子錯体	E (KJ mol^{-1})
CTAB／2－メチルインドール	－4.155
MTAB／2－メチルインドール	－3.265
LTAB／2－メチルインドール	－2.429
CTAB／スカトール	－3.858
MTAB／スカトール	－3.041
LTAB／スカトール	－2.215

If the die length of an alkyl chain decreases so that drawing 9 , drawing 11 , etc. may see, the number of contact of the alkyl chain located in less than 5A will decrease. This is van so that clearly from Table 5. der It is based on reduction of waals energy. Moreover, vander The molecule complex of large 2-methylindole of waals energy is incorporated by the surfactant crystal lattice from skatole at stability, and it has explained that an evaporation rate was slow and it was extremely stable.

[0044] As mentioned above, molecule complex crystal formation with a surfactant and an aromatic compound can control the evaporation rate with the die length of the chain of the surfactant which delays the evaporation rate of an aromatic compound and is used.

[0045]

[Effect of the Invention] The crystal and approach by this invention suppress evaporation of an aromatic compound, for example, an aromatic and an insecticide, the charge of makeup, physic, etc. simple and cheaply, and sustained-release can be given and they can adjust the gradual release operation easily.

[Translation done.]

* NOTICES *

DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]
[Drawing 1] It is a a-axis projection drawing and b b-axis projection drawing of the crystal structure of crystal 1-I.
[Drawing 2] It is drawing showing the molecular structure of crystal 1-I.
[Drawing 3] It is the b-axis projection drawing of the crystal structure of crystal 1-II.
[Drawing 4] It is drawing showing the molecular structure of crystal 1-II.
[Drawing 5] It is the c-axis projection drawing of the crystal structure of crystal 1-III.
[Drawing 6] It is drawing showing the molecular structure of crystal 1-III.
[Drawing 7] It is the graph which shows the result of a dissolution rate measurement experiment.

[Drawing 8] It is the graph which shows the result of division nature and a stability measurement experiment.
[Drawing 9] It is a a-axis projection drawing and b b-axis projection drawing of the crystal structure of crystal 2-I.
[Drawing 10] It is drawing showing the molecular structure of crystal 2-I.
[Drawing 11] They are crystal 2-II(a) and the b-axis projection drawing of the crystal structure of III (b).
[Drawing 12] It is drawing showing the molecular structure of crystal 2-II(a) and III (b).
[Drawing 13] It is the b-axis projection drawing of the crystal structure of crystal 2-IV.
[Drawing 14] They are crystal 2-IV(a), V (b), and drawing showing the molecular structure of VI (c).
[Drawing 15] It is drawing showing the molecular structure of crystal 2-VII.
[Drawing 16] It is the graph which shows the result of sustained-release and the stability measurement experiment conducted by 2-methylindole.
[Drawing 17] It is the graph which shows the result of sustained-release and the stability measurement experiment conducted by skatole.

[Translation done.]